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THEORETICAL CONSIDERATIONS REGARDING THE GROWTH AND DECAY OF THE SHORT-LIVED DECAY PRODUCTS OF RADON AND THORON PLATED-OUT ON LARGE SURFACES

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by

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ABSTRACT

The growth of activity, differential and integrated, of the short-lived decay products of radon and thoron plated-out on large surfaces during exposure, and the decay of activity of the above radioactive products after exposure, have been investigated. Analytical expressions are given which are suitable for the calculation of deposition velocities, and other variables, from the experimental determination of the surface activity on a given exposed material surface. Other possible applications to personal dosimetry and environmental monitoring suggest themselves.

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OBSERVATIONS THÉORIQUES SUR LA CROISSANCE ET LA DÉSINTÉGRATION DES PRODUITS DE DÉSINTÉGRATION À COURTE VIE DU RADON ET DU THORON DÉPOSÉS SUR DE GRANDES SURFACES

par

J. Bigu*

RÉSUMÉ

On a fait l'étude de la croissance de l'activité, différentielle et intégrée, des produits de désintégration à courte vie du radon et du thoron déposés sur de grandes surfaces lors de l'exposition ainsi que de la désintégration de l'activité des produits radioactifs décrits ci-dessus après exposition. Les expressions analytiques fournies conviennent au calcul des vitesses de déposition et des autres variables tirées de la détermination expérimentale de l'activité de surface d'une surface déterminée de matière exposée. Enfin, d'autres applications possibles à la dosimétrie personnelle et au contrôle de l'environnement viennent à l'esprit d'elles-mêmes.

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INTRODUCTION

The short-lived decay products of radon and thoron are initially formed in an atomic, positively charged, state of great diffusivity. These decay products readily attach themselves to small surfaces such as aerosols and to large surfaces such as walls. Attachment of particles to large surfaces is commonly referred to as plate-out. The degree of plate-out depends on environmental conditions including aerosol concentration, air moisture content, the presence of tracer gases, and most probably on the properties of the surface. Although theoretical and experimental data are available on the attachment of the radon and thoron progeny to aerosols (1-11), less is known on the attachment to walls, particularly with regard to the chemical nature and surface physical characteristics of the walls.

Plate-out of the radon and thoron progeny on walls and other objects is a problem of great practical interest and concern when evaluating radiation levels by environmental monitors and personal dosimeters. This is so because plate-out affects the accuracy and reliability of the results obtained by some radiation instrumentation as decay products of radon and thoron may be removed by the 'sampling heads' and other components of the instrument. Plate-out of these decay products is also relevant in connection with air quality control in underground uranium mines, mills, and other uranium related industries, and in the operation of radon (thoron) test facilities for simulation studies, calibration of instrumentation and for other practical purposes.

Data on the rate of attachment (plate-out) of radioactive particles on walls are used to calculate the particle deposition velocity. Both variables, i.e., attachment rate and deposition velocity, are, in turn, used to estimate the extent of plate-out on sampling heads, and to evaluate the effect of large surfaces on radiation levels in specific working areas and test

facilities, as indicated above.

Deposition rates are determined experimentally by measuring the surface activity on some samples designed for the purpose. These samples usually assume the form of small 'discs' which are attached to the surface where plate-out is to be determined. The samples are used because plate-out measurements carried out directly on the surface of interest may not be practical, and in most cases quite difficult.

This paper deals with the growth and decay of the decay products of radon and thoron plated-out on large surfaces. The paper gives analytical expressions suitable for the calculation of deposition velocities, and other variables, from the experimental determination of surface activity on a given material surface.

THEORETICAL BACKGROUND

The deposition velocity, V, is defined by the following relationship:

$$V = \frac{Y}{N} \qquad \text{Eq 1}$$

where, Y is the deposition (attachment) rate, i.e., number of particles plated-out per unit surface area of wall per unit time; and N is the number of particles per unit volume.

The deposition rate for a given radioactive product, say m, can be expressed as follows:

$$Y_m = f_m V_m C_m \qquad \text{Eq 2}$$

where C_m is the concentration of decay product m, f_m stands for the attached or unattached fraction of product m. Because the deposition rate of attached decay products is considerably smaller than that corresponding to the unattached fraction on account of the much lower diffusivity of the former as compared with the latter, the symbol f will preferentially be used to denote the unattached

fraction.

The growth of plated-out activity on a given surface exposed to a radon/radon progeny atmosphere, thoron/thoron daughter atmosphere, or an arbitrary mixture of the above, is given by the following expression:

$$\frac{dS_m}{dt} = C_m f_m V_m + S_{m-1}\,\lambda_m - S_m \lambda_m \qquad \text{Eq 3}$$

where, the symbols S_m and S_{m-1} stand for surface activity, say Bq/m^2 or $atoms/m^2$, corresponding to atoms of type m, and to parent atom m-1, respectively. The symbol λ stands for radioactive decay constant.

It should be noted that the first and second terms on the right hand side of Eq. 3 represent the growth of m on the surface by direct plate-out (first term), and by decay of its parent product, respectively. The third term of the above Equation represents the removal of product m by radioactive decay.

Using the indices i, j and k to denote, respectively, RaA or ThA, RaB or ThB and RaC or ThC, one may write Eq. 3 for the decay products as follows:

$$\frac{dS_i}{dt} = C_i f_i V_i - S_i \lambda_i \qquad \text{Eq 4}$$

$$\frac{dS_j}{dt} = C_j f_j V_j + S_i \lambda_j - S_j \lambda_j \text{ , and} \qquad \text{Eq 5}$$

$$\frac{dS_k}{dt} = C_k f_k V_k + S_j \lambda_k - S_k \lambda_k \qquad \text{Eq 6}$$

The solutions of Equations 4 to 6 are attained by standard methods of differential calculus. It should be noted that the solution of Eq. 4 for S_i is then used in Eq. 5 to solve for S_j, and so on. Hence, the calculation increases considerably in complexity as the analytical procedure proceeds from Eq. 4 to Eq. 6.

The solutions of Equations 4 to 6 are given below:

$$S_i(T_s) = \frac{C_i f_i V_i}{\lambda_i}\,(1 - e^{-\lambda_i T_s}) \qquad \text{Eq 7}$$

$$S_j(T_s) = \frac{C_j f_j V_j}{\lambda_j}\left(1 - e^{-\lambda_j T_s}\right) + C_i f_i V_i \lambda_j \left[\frac{1 - e^{-\lambda_i T_s}}{\lambda_i(\lambda_j - \lambda_i)} + \frac{1 - e^{-\lambda_j T_s}}{\lambda_j(\lambda_i - \lambda_j)}\right] \qquad \text{Eq 8}$$

$$S_k(T_s) = \frac{C_k f_k V_k}{\lambda_k}\left(1 - e^{-\lambda_k T_s}\right) + C_j f_j V_j \lambda_k \left[\frac{1 - e^{-\lambda_j T_s}}{\lambda_j(\lambda_k - \lambda_j)} + \frac{1 - e^{-\lambda_k T_s}}{\lambda_k(\lambda_j - \lambda_k)}\right]$$

$$+ C_i f_i V_i \lambda_j \lambda_k \left[\frac{1 - e^{-\lambda_i T_s}}{\lambda_i(\lambda_j - \lambda_i)(\lambda_k - \lambda_i)} + \frac{1 - e^{-\lambda_j T_s}}{\lambda_j(\lambda_i - \lambda_j)(\lambda_k - \lambda_j)} + \frac{1 - e^{-\lambda_k T_s}}{\lambda_k(\lambda_i - \lambda_k)(\lambda_j - \lambda_k)}\right] \qquad \text{Eq 9}$$

The steady-state condition for the activities accumulated on the surface of the material is obtained for a sampling time T_s much longer than the half-life ($T_{1/2}$) of the radioisotope of interest. This condition is expressed as follows:

$$\lambda_m T_s = 0.693 T_s / T_{1/2}(m) \gg 1, \text{ or } e^{-\lambda_m T_s} \cong 0,$$

or $$1 \gg e^{-\lambda_m T_s} \quad \text{where } m = i, j, \text{ or } k.$$

Taking into account that:

$$\{1/\lambda_i(\lambda_j - \lambda_i)\} + \{1/\lambda_j(\lambda_i - \lambda_j)\} = 1/\lambda_i\lambda_j.$$

$$\{1/\lambda_j(\lambda_k - \lambda_j)\} + \{1/\lambda_k(\lambda_j - \lambda_k)\} = 1/\lambda_j\lambda_k,$$

and

$$\{1/\lambda_i(\lambda_j - \lambda_i)(\lambda_k - \lambda_i)\} + \{1/\lambda_j(\lambda_i - \lambda_j)(\lambda_k - \lambda_j)\} + \{1/\lambda_k(\lambda_i - \lambda_k)(\lambda_j - \lambda_k)\} = 1/\lambda_i\lambda_j\lambda_k,$$

It is not difficult to show that the steady-state surface activities $S_{i,o}(T_s)$, $S_{j,o}(T_s)$ and $S_{k,o}(T_s)$, where the sub-index o is used to indicate steady-state, are given by:

$$S_{i,o}(T_s) = C_i f_i V_i / \lambda_i \qquad \text{Eq 10}$$

$$S_{j,o}(T_s) = (C_i f_i V_i/\lambda_i) + (C_j f_j V_j/\lambda_j) = S_{i,o}(T_s) + (C_j f_j V_j/\lambda_j) \qquad \text{Eq 11}$$

$$S_{k,o}(T_s) = (C_i f_i V_i/\lambda_i) + (C_j f_j V_j/\lambda_j) + (C_k f_k V_k/\lambda_k)$$

$$= S_{j,o}(T_s) + C_k f_k V_k/\lambda_k \qquad \text{Eq 12}$$

It should be noted that the same results can be obtained by setting $dS_i/dt = dS_j/dt = dS_k/dt = 0$, and solving Equations 4 to 6.

The total surface activities accumulated on the surface during an

exposure or sampling time T_{s2} to T_{s1} ($T_{s2}>T_{s1}$) are obtained by integrating Equations 7, 8 and 9. Calling these integrated activities IS_iS, IS_jS, and IS_kS, where I stands for integrated and S for sampling, it is not difficult to show that:

$$IS_iS = \int_{T_{s1}}^{T_{s2}} S_i(T_s)\, dT_s = C_i f_i V_i \tau_i F_i \qquad \text{Eq 13}$$

$$IS_jS = \int_{T_{s1}}^{T_{s2}} S_j(T_s) dT_s = C_j f_j V_j \tau_j F_j + C_i f_i V_i \left(\frac{\tau_i^2 F_i}{\tau_i - \tau_j} + \frac{\tau_i \tau_j F_i}{\tau_j - \tau_i} \right) \qquad \text{Eq 14}$$

and,

$$IS_kS = \int_{T_{s1}}^{T_{s2}} S_k(T_s) dT_s = C_k f_k V_k \tau_k F_k + C_j f_j V_j \left(\frac{\tau_j^2 F_j}{\tau_j - \tau_k} + \frac{\tau_j \tau_k F_k}{\tau_k - \tau_j} \right) +$$

$$+ C_i f_i V_i \left(\frac{\tau_i^3 F_i}{(\tau_i - \tau_j)(\tau_i - \tau_k)} + \frac{\tau_i \tau_j^2 F_j}{(\tau_j - \tau_i)(\tau_j - \tau_k)} + \frac{\tau_i \tau_k^2 F_k}{(\tau_k - \tau_i)(\tau_k - \tau_j)} \right) \qquad \text{Eq 15}$$

where,

$\tau_i = 1/\lambda_i$, $\tau_j = 1/\lambda_j$ and $\tau_k = 1/\lambda_k$.

$F_i = (T_{s2} - T_{s1}) + \tau_i (e^{-T_{s2}/\tau_i} - e^{-T_{s1}/\tau_i})$

$F_j = (T_{s2} - T_{s1}) + \tau_j (e^{-T_{s2}/\tau_j} - e^{-T_{s1}/\tau_j})$

$F_k = (T_{s2} - T_{s1}) + \tau_k (e^{-T_{s2}/\tau_k} - e^{-T_{s1}/\tau_k})$

If the exposed surface is now removed from the radiation site to a location of negligible radiation level after a sampling period, T_s, in the former, the activities remaining on the surface at a time T_w after the end of the sampling period, $S_m(T_w)$, can be obtained from Equation 3 by setting the first term on the right hand side equal to zero, as $C_m=0$. Furthermore, for the first product of the radioactive series, i.e., RaA and ThA, the second term on the right hand side of Equation 3 also reduces to zero. Using Equations 4 to 6, taking into account the above, it is easy to show that:

$$S_i(T_w) = S_i(T_s) e^{-\lambda_i T_w} \qquad \text{Eq 16}$$

$$S_j(T_w) = S_j(T_s)\, e^{-\lambda_j T_w} + S_i(T_s)\lambda_j\left[\frac{e^{-\lambda_i T_w}}{\lambda_j-\lambda_i} + \frac{e^{-\lambda_j T_w}}{\lambda_i-\lambda_j}\right] \qquad \text{Eq 17}$$

$$S_k(T_w) = S_k(T_s)\, e^{-\lambda_k T_w} + S_j(T_s)\lambda_k\left(\frac{e^{-\lambda_j T_w} - e^{-\lambda_k T_w}}{\lambda_k - \lambda_j}\right) + S_i(T_s)\lambda_i\lambda_k$$

$$\left[\frac{e^{-\lambda_i T_w}}{(\lambda_j-\lambda_i)(\lambda_k-\lambda_i)} + \frac{e^{-\lambda_j T_w}}{(\lambda_i-\lambda_j)(\lambda_k-\lambda_j)} + \frac{e^{-\lambda_k T_w}}{(\lambda_i-\lambda_k)(\lambda_j-\lambda_k)}\right] \qquad \text{Eq 18}$$

where $S_i(T_s)$, $S_j(T_s)$ and $S_k(T_s)$ are given, respectively, by Equations 7, 8 and 9.

During the waiting period, the activities on the surface of the material diminish with increasing time. The integrated surface activity during a waiting period T_{w1} to T_{w2} ($T_{w2}>T_{w1}$) can be obtained by integrating Equations 16, 17 and 18 during the waiting period. Calling IS_iW, IS_jW and IS_kW the integrated surface activities over the waiting period it is not difficult to show that:

$$IS_iW = \int_{T_{w1}}^{T_{w2}} S_i(T_w)dT_w = C_i f_i V_i \tau_i^2 \phi_i \qquad \text{Eq 19}$$

$$IS_jW = \int_{T_{w1}}^{T_{w2}} S_j(T_w)dT_w = C_i f_i V_i\left(\frac{\tau_i^3\phi_i}{\tau_i-\tau_j} + \frac{\tau_j^2\tau_i\phi_j}{\tau_j-\tau_i}\right) + C_j f_j V_j \tau_j^2 \phi_j \qquad \text{Eq 20}$$

$$IS_kW = \int_{T_{w1}}^{T_{w2}} S_k(T_w)dT_w$$

$$= C_i f_i V_i\left[\frac{\tau_i^4\phi_i}{(\tau_i-\tau_j)(\tau_i-\tau_k)} + \frac{\tau_j^3\tau_i\phi_j}{(\tau_j-\tau_i)(\tau_j-\tau_k)} + \frac{\tau_k^3\tau_i\phi_k}{(\tau_k-\tau_i)(\tau_k-\tau_j)}\right]$$

$$+ C_j f_j V_j\left[\frac{\tau_j^3\phi_j}{\tau_j-\tau_k} + \frac{\tau_k^2\tau_j\phi_k}{\tau_k-\tau_j}\right] + C_k f_k V_k \tau_k^2 \phi_k \qquad \text{Eq 21}$$

where, $\phi_i = (1 - e^{-T_{s2}/\tau_i})(e^{-T_{w1}/\tau_i} - e^{-T_{w2}/\tau_i})$

$\phi_j = (1 - e^{-T_{s2}/\tau_j})(e^{-T_{w1}/\tau_j} - e^{-T_{w2}/\tau_j})$

$\phi_k = (1 - e^{-T_{s2}/\tau_k})(e^{-T_{w1}/\tau_k} - e^{-T_{w2}/\tau_k})$

PRACTICAL APPLICATIONS

Equations 10 to 12 can be used to determine an average or 'overall' deposition velocity for the radon (thoron) progeny if the assumption is made that $V_i = V_j = V_k$ provided C_i, C_j and C_k, and f_i, f_j and f_k are known. These variables should be determined during the sampling or exposure period by standard methods designed for this purpose and available in the pertinent literature. The deposition velocity can then easily be determined from measurements of, say, the total α-surface activity as indicated by the author elsewhere (10,11).

However, there is inadequate evidence to support the statement of equality of the deposition velocity for the different decay products, as indicated above. For the more rigorous and general case where no assumptions are made regarding the deposition velocities of members of the same decay chain, the determination of V_i, V_j and V_k becomes substantially more complicated, experimentally and analytically. For this more general case, use of Equations 19 to 21 is made.

Equations 19, 20 and 21 can be rewritten, for simplicity, as follows:

$$IS_iW = AV_i \qquad \text{Eq 22}$$

$$IS_jW = BV_i + DV_j \qquad \text{Eq 23}$$

$$IS_kW = EV_i + GV_j + HV_k \qquad \text{Eq 24}$$

where,

$$A = C_i f_i \tau_i^2 \phi_i$$

$$B = C_i f_i \left(\frac{\tau_i^3 \phi_i}{\tau_i - \tau_j} + \frac{\tau_j^2 \tau_i \phi_j}{\tau_j - \tau_i} \right)$$

$$D = C_j f_j \tau_j^2 \phi_j$$

$$E = C_i f_i \left[\frac{\tau_i^4 \phi_i}{(\tau_i - \tau_j)(\tau_i - \tau_k)} + \frac{\tau_j^3 \tau_i \phi_j}{(\tau_j - \tau_i)(\tau_j - \tau_k)} + \frac{\tau_k^3 \tau_i \phi_k}{(\tau_k - \tau_i)(\tau_k - \tau_j)} \right]$$

$$G = C_j f_j \left[\frac{\tau_j^3 \phi_j}{\tau_j - \tau_k} + \frac{\tau_k^2 \tau_j \phi_k}{\tau_k - \tau_j} \right]$$

$$H = C_k f_k \tau_k^2 \phi_k$$

Determination of V_i, V_j and V_k from Equations 22 to 24 can be done by measuring the gross α-activity, or by α-spectroscopy, on the surface of the material. Other methods using β-counting, and γ-counting and γ-spectroscopy are also possible.

If gross α-count is used, the total α-count, $N_{\alpha,T}$, recorded by a detector with α-counting efficiency ε is given by:

$$N_{\alpha,T} = 2.22\varepsilon(IS_iW + IS_kW)$$

$$= 2.22\varepsilon[V_i(A+E) + GV_j + HV_k] \qquad \text{Eq 25}$$

Because there are three unknowns, at least three independent α-measurements have to be done on the surface of the material after exposure, in order to determine V_i, V_j and V_k.

There are a variety of optimized α-counting schemes available in the literature (12-27) that can be used in conjunction with Equations 22 to 25 to determine, after some algebraic manipulation, the variables of interest, namely V_i, V_j and V_k.

It should be noted that the growth of activity and integrated activity deposited by plate-out on a given surface, described by Equations 7, 8 and 9, and Equations 13, 14 and 15, respectively, are similar to that obtained on a filter through which air is sampled by means of a pump with a sampling rate Q. Comparison can be made between these two cases by substituting the rate of decay product deposition on the filter by active sampling, denoted by QC_m, by the attachment rate due to plate-out, namely: $C_m f_m V_m$, in the relevant kinetic differential equations. As the problem of active sampling under steady-state and time-dependent conditions has been discussed in great detail by the author

elsewhere (27-29), the dynamics of Equations 7 to 9 and 13 to 15 readily follow, and will therefore not be pursued here any further. The interested person should refer to the above references for more information.

The equations developed in this work could also find application in the fields of personal dosimetry and environmental monitoring.

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